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ANDREA DeGUCHI  
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Date

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

## IN RE APPLICATION OF

Bernhard Müller

Group Art Unit: M. Einsmann

APPLICATION NO: 08/801,327

Examiner: 1751

FILED: February 18, 1997

FOR: FIBRE REACTIVE ANTHRAQUINONE  
DYES, PROCESS FOR THEIR  
PREPARATION AND THE USE THEREOF

Assistant Commissioner for Patents

Washington, D.C. 20231

TRANSMITTAL LETTER

Sir:

Enclosed herewith are three copies of the Appeal Brief in the above-identified application.

- Please charge Deposit Account No. 03-1935 in the amount of \$300.00 for payment of the fee. Two additional copies of this paper are here enclosed. The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment, to Account No. 03-1935.

Respectfully submitted,

Kevin T. Mansfield  
Agent for Applicant  
Reg. No. 31,635

Ciba Specialty Chemicals Corporation  
Patent Department  
540 White Plains Road  
P.O. Box 2005  
Tarrytown, NY 10591-9005  
(914) 785-7127  
KTM/am/

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

IN RE APPLICATION OF

Group Art Unit: 1751

BERNHARD MÜLLER

Examiner: M. Einsmann

APPLICATION NO: 08/801,327

FILED: FEBRUARY 18, 1997

FOR: FIBRE-REACTIVE ANTHRAQUINONE

DYES, PROCESS FOR THEIR  
PREPARATION AND THE USE THEREOF

Assistant Commissioner for Patents

Washington, D.C. 20231

APPEAL BRIEF

Sir:

This appeal is from the final rejection of claims 2, 3, 5-10 and 16-19 mailed from the PTO on September 24, 1999.

The Notice of Appeal was mailed to the Patent and Trademark Office by first class mail with Certificate of Mailing on December 22, 1999. The Notice of Appeal was date stamped in the PTO Mail Room on December 27th, making this Brief due on Sunday, February 27, 2000.

(1) REAL PARTY OF INTEREST

The real party of interest is:

Ciba Specialty Chemicals Corp.  
P.O. Box 2005  
560 White Plains Road  
Tarrytown, New York 10591

(2) RELATED APPEALS AND INTERFERENCES

To the knowledge of the undersigned, there are no related appeals and/or interferences.

(3) STATUS OF THE CLAIMS

Claims 1, 4 and 13-15 have been cancelled. No claims have been withdrawn.

According to the final rejection mailed from the PTO on September 24, 1999, claims 11-12 are allowed. The indication in the subsequent Advisory Action that they remain objected to due to dependency is believed to be in error since claim 11 was previously rewritten in independent form, with claim 12 dependent thereon. In view of this controversy, claims 11-12 remain in the appended claims.

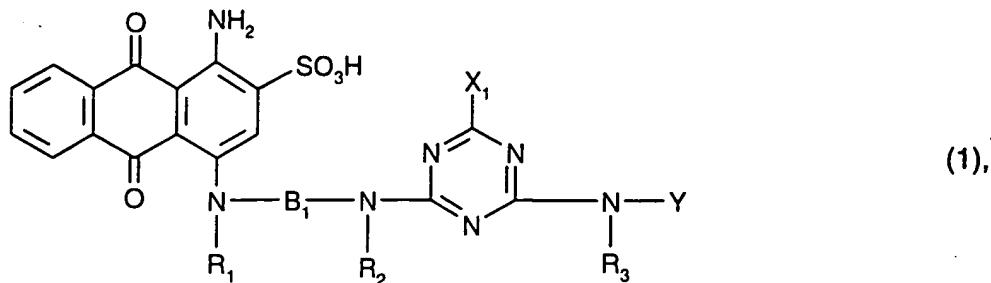
Claims 2, 3, 5-10 and 16-19 stand finally rejected. The omission of mention of claim 19 in the subsequent Advisory Action is also believed to be in error since it had been previously rejected and was still pending.

(4) STATUS OF AMENDMENTS

The claims were not further amended in response to the final rejection.

## (5) SUMMARY OF THE INVENTION

The presently claimed invention relates to novel, reactive anthraquinone dyes of the formula



wherein R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> are each independently of one another hydrogen or C<sub>1</sub>-C<sub>12</sub>alkyl which is unsubstituted or substituted by hydroxyl, sulfo or sulfato and, with the exception of methyl, may be interrupted by oxygen (page 1, last 5 lines to page 2, line 16), X<sub>1</sub> is chloro or fluoro (page 2, line 3), B<sub>1</sub> is methylene-phenylene-methylene which is unsubstituted or substituted in the phenylene ring by C<sub>1</sub>-C<sub>4</sub>alkyl, C<sub>1</sub>-C<sub>4</sub>alkoxy, C<sub>2</sub>-C<sub>4</sub>alkanoylamino, halogen, carboxy or sulfo, or is a radical of the formula -CH<sub>2</sub>-CH<sub>2</sub>-CH(C<sub>2</sub>H<sub>5</sub>)-, -CH<sub>2</sub>-CH(OH)-CH<sub>2</sub>- or -CH<sub>2</sub>-C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>- (page 2, lines 4-7), Y is hydrogen, or C<sub>1</sub>-C<sub>12</sub>alkyl which is unsubstituted or substituted by hydroxyl, sulfo or sulfato and, with the exception of methyl, may be interrupted by oxygen, or phenyl or naphthyl, each unsubstituted or substituted by C<sub>1</sub>-C<sub>4</sub>alkyl, C<sub>1</sub>-C<sub>4</sub>alkoxy, C<sub>2</sub>-C<sub>4</sub>alkanoylamino, halogen, carboxy, sulfo or a radical of formula -SO<sub>2</sub>-Z, wherein Z is a group of formula -CH=CH<sub>2</sub> or -CH<sub>2</sub>-CH<sub>2</sub>-U<sub>1</sub>, and U<sub>1</sub> is a leaving group (page 2, lines 8-16), or Y is an anthraquinone of the formula



wherein B<sub>2</sub> is C<sub>2</sub>-C<sub>12</sub>alkylene which is unsubstituted or substituted by hydroxyl, sulfo or sulfato, and which may be interrupted by oxygen, or methylene-phenylene-methylene which is unsubstituted or substituted in the phenylene ring by C<sub>1</sub>-C<sub>4</sub>alkyl, C<sub>1</sub>-C<sub>4</sub>alkoxy, C<sub>2</sub>-C<sub>4</sub>alkanoylamino, halogen, carboxy or sulfo (page 2, lines 17-19 and page 3, lines 17-22), as claimed in independent claim 16.

Claim 2 is directed to the preferred genus disclosed in the third and fourth full paragraphs of page 3.

Claim 3 is directed to the more preferred small genus disclosed in the sixth full paragraph of page 3.

Claim 5 is directed to the preferred genus disclosed in the second paragraph of page 5. Claim 6 is

directed to the particularly preferred genus disclosed on page 2, lines 20-25 and page 4, lines 3-4. Claim 7 is directed to the more particularly preferred genus disclosed on page 4, lines 9-10. Claim 8 is directed to the most particularly preferred genus disclosed on page 4, line 11. Claim 9 is directed to the preferred genus disclosed in the fourth full paragraph of page 5. Claim 10 is directed to the more preferred genus disclosed in the second paragraph of page 6. Claim 11 is directed to the preferred genus disclosed in the first paragraph of page 6. Claim 12 is directed to the more preferred genus disclosed in the second paragraph of page 6. Claims 17 and 18 are supported by the disclosure in the first and second full paragraphs of page 8. Claim 19 is directed to the most particularly preferred embodiment of the genus disclosed in the second paragraph of page 6 as well as by Example 7 on page 12.

As taught in the first and second full paragraphs of page 8, the novel, reactive anthraquinone dyes of the present invention are suitable for dyeing or printing a wide variety of hydroxyl-group-containing or nitrogen-containing fiber materials such as cellulosic fiber materials like cotton and regenerated cellulose or natural or synthetic polyamide fiber materials such as wool, silk and synthetic polyamide. According to the last full paragraph of page 8 and the first paragraph of page 9, the novel, reactive anthraquinone dyes of the present invention are distinguished by high reactivity, good fixation and very good buildup. They also have very good all-around fastness properties including good fastness to light and wet processing.

#### (6) ISSUES

Only one issue is presented for review, it being assumed that the objection to claims 11 and 12 due to dependency is in error:

Whether claims 2, 3, 5-10 and 16-19 are unpatentable under 35 U.S.C. § 103(a) over Harms, GB 2,034,731.

#### (7) GROUPING OF THE CLAIMS

The claims do not stand or fall together. Claims 8-10 and 19 of narrower scope are argued separately.

(8) ARGUMENT

Harms, GB 2,034,731, generically discloses water soluble reactive dyes which include the dyes of instant formula (1) when the diamino bridging member connecting the anthraquinone chromophore and the triazine ring (= A in Harms, B<sub>1</sub> in the claims) is an aliphatic bridging member. Harms teaches that the bridging member may be a straight or branched chain alkylene having 2-15 carbon atoms. Preferably it is a straight or branched C<sub>2</sub>-C<sub>8</sub>alkylene chain.

Harms teaches that his dyes of formula (I) are suitable for dyeing or printing textile materials containing hydroxyl groups or nitrogen groups such as natural and regenerated cellulose and wool, silk and synthetic polyamide. The dyeings are taught to have good fastness properties to light and wet processing.

While Harms discloses a number of bridging members on pages 1-2, no dye containing an alkylene bridging member of the formula -CH<sub>2</sub>-CH<sub>2</sub>-CH(C<sub>2</sub>H<sub>5</sub>)-, -CH<sub>2</sub>-CH(OH)-CH<sub>2</sub>- or -CH<sub>2</sub>-C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>- as presently claimed is disclosed. Further, the possibility of hydroxyl substitution as in the bridging member of the formula -CH<sub>2</sub>-CH(OH)-CH<sub>2</sub>- is unsuggested.

Thus the exemplified dyes of Harms differ from the instantly claimed dyes of generic claim 16 with respect to the bridge member. With regard to the examiner's assertion that it is clear from the disclosure that 5 and 6 carbon isomers are preferred embodiments, pointing to Harms's disclosure starting at page 1, line 32 through page 2, appellant notes that 6 carbon isomers are no longer claimed. Moreover, since over 90% of the preferred bridging members in the cited section of Harms are not 5 carbon isomers, it is difficult to ascertain any preference for C<sub>5</sub>alkylene. Nevertheless, appellant does not dispute that the alkylene bridging members claimed herein of the formulae -CH<sub>2</sub>-CH<sub>2</sub>-CH(C<sub>2</sub>H<sub>5</sub>)- and -CH<sub>2</sub>-C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>- would have been obvious to the skilled artisan absent a showing of criticality. Appellant acknowledges that the preferred bridge member of claims 8, 9, 10 and 19 is specifically listed on line 35 of page 1 of Harms, but points out it does not appear in any of the disclosed dyes.

In response to the claims rejection, the examiner was requested to consider the results of the comparative tests in the declaration dated August 22, 1996 (copy enclosed for the Board's convenience) by Dr. Bernard Müller, the named inventor.

In said enclosed Declaration the dye of Example 78 on page 18 of Harms was compared with an instantly claimed dye. Dyestuff A corresponds to the single dye claimed in claim 19. It is within the small genus of dyes having the same diamino bridge member claimed in claims 8, 9 and 10. Dyestuff X is the dye of Example 78 of Harms. The dyestuffs A and X in the declaration only differ from each other with respect to the diamino bridge member, the inventive compound having a 3 carbon chain between the aromatic rings, with 2 methyl groups on one of the carbons and the reference compound having a 3 carbon chain between the aromatic rings, with one methyl group on one of the nitrogens.

The declaration clearly demonstrates that the dye according to the present application has a distinctly better buildup, and hence better affinity to cotton fiber material. The dye expert averred that the extent of the improvement was very surprising and of commercial importance, and he would not have predicted it.

The rejection of all the appealed claims was maintained, with the examiner pointing to the diamino bridge member of Examples 48 and 74 of Harms. Appellant acknowledges that the dyes of the cited reference having the closest structural similarity to the instantly claimed dyestuffs are the dyes obtainable from the reactants shown in Examples 48 and 74 of Harms. Accordingly, a proper comparison with the dyes of the prior art should be made with the dyes of said Examples. Said Examples are both based on the same C<sub>5</sub>-diamine, 2,4-diaminopentane.

Since 2,4-diaminopentane is not commercially available, at least according to appellant's knowledge, and the reference is absolutely silent on how it is obtainable, appellant tried to synthesize said diamine according to procedures known in the art of amine preparation. Two approaches were taken which were diligently pursued over a period of 20 months. These approaches were: (i) catalytic hydrogenation of pentanone-2,4-dioxime and (ii) reductive amination of 2,4-dioxopentane. The details of these pursuits are provided in the declaration dated July 7, 1999 (copy enclosed for the Board's convenience) by Dr. Bernard Müller.

With approach (i), three different hydrogenation catalysts were tried. However, each of these efforts to synthesize 2,4-diaminopentane failed. Likewise approach (ii) also failed. Therefore, the present declaration at least demonstrates that the teaching of the prior art fails to satisfy the enablement requirement with regard to the dyes of Examples 48 and 74, which is indispensable for a disclosure to constitute prior art against any later invention.

Appellant notes that no significant properties are listed for the dyes of Examples 48 and 74. Further, from page 11, lines 28-30 and page 17, lines 27-28, it appears that Examples 48 and 74 (as well as the other dyes obtainable if the indicated reactants are employed), are merely prophetic. Since 2,4-diaminopentane is not commercially available and appellant has shown it cannot be made by procedures known in the art of amine preparation, it is averred that one skilled in the art was not enabled to make and use the invention disclosed in Examples 48 and 74 in the reference at the time the instant invention was made.

It is well established that the mere naming of a compound in a reference without more cannot constitute a description of a compound, particularly when the evidence of record suggests that no method exists for its preparation. See *In re Wiggins et al.*, 179 USPQ 421, 425 (CCPA, 1973). See also *In re Hoeksema*, 399 F.2d 269, 274, 158 USPQ 596, 601 (CCPA 1968).

The examiner initially responded by stating that the validity of a patent cannot be questioned. This statement is believed to be incorrect in fact and law. Patents may not only be questioned; they can be and in many cases are invalidated, either in litigation or by the PTO during interference, reissue, reexamination and public use proceedings.

According to the Manual of Patent Examining Procedure (MPEP), § 2121 a reference is presumed to be operable. The burden then shifts to the applicant to provide facts rebutting the presumption of operability. See *In re Sasse*, 629 F.2d 675, 207 USPQ 107 (CCPA 1980).

MPEP § 2121 further points out that it does not matter whether the prior art reference is a U.S. patent, a foreign patent, a printed publication or other. The level of disclosure required to make reference an "enabling disclosure" is the same. There is no basis in the statute (35 U.S.C. 102 or 103) for discriminating either in favor of or against prior art references on the basis of nationality.

MPEP § 2121.01 states that a reference contains an "enabling disclosure" if the public was in possession of the claimed invention before the date of invention. "Such possession is effected if one of ordinary skill in the art could have combined the publication's description of the invention with his [or her] own knowledge to make the claimed invention". *In re Donohue*, 766 F.2d 531, 226 USPQ 619 (CAFC, 1985).

As set forth in MPEP § 2121.02, "a reference is presumed operable until applicant provides facts rebutting the presumption of operability. *In re Sasse*, 629 F.2d 675, 207 USPQ 107 (CCPA 1980). Therefore, applicant must provide evidence showing that a process for making was not known at the time of the invention".

"When a prior art reference merely discloses the structure of the claimed compound, evidence showing that attempts to prepare that compound were unsuccessful before the date of invention will be adequate to show inoperability. *In re Wiggins*, 488 F.2d 538, 179 USPQ 421 (CCPA 1971)".

Appellant also notes the Guidelines for the Examination of Claims Directed to Species of Chemical Compositions Based Upon a Single Prior Art Reference, published by Bruce A. Lehman, Assistant Secretary of Commerce and Commissioner of Patents and Trademarks in September of 1998. Its summary states

The Patent and Trademark Office (PTO) is publishing the final version of guidelines to be used by Office personnel in reviewing a certain type of patent application for compliance with 35 U.S.C. 103. The guidelines are to be used when examining claims directed to a species or subgenus of chemical compositions when: (1) the claims have been rejected based upon a single prior art reference, and (2) the single prior art reference discloses a genus embracing the claimed species or subgenus but does not expressly describe the particular claimed species or subgenus.

According to footnote 55 therein, the court in *In re Hoeksema* stated:

"Thus, upon careful reconsideration it is our view that if the prior art of record fails to disclose or render obvious a method for making a claimed compound, at the time the invention was made, it may not be legally concluded that the compound itself is in the possession of the public. [footnote omitted.] In this context, we say that the absence of a known or obvious process for making the claimed compounds overcomes a presumption that the compounds are obvious, based on close relationships between their structures and those of prior art compounds." [Emphasis added]

The *Hoeksema* court further noted that once a *prima facie* case of obviousness is made by the PTO through citation of references, the burden is on the applicant to produce contrary evidence establishing that the reference being relied on would not enable a skilled artisan to produce the

different compounds claimed. Id. at 274-75, 158 USPQ at 601. See also *Ashland Oil, Inc. v. Delta Resins & Refractories, Inc.*, 776 F.2d 281, 295, 297, 227 USPQ 657, 666, 667 (Fed. Cir. 1985) (citing *Hoeksema* for the proposition above); *In re Grose*, 592 F.2d 1161, 1168, 201 USPQ 57, 63-64 (CCPA 1979) ("One of the assumptions underlying a *prima facie* obviousness rejection based upon a structural relationship between compounds, such as adjacent homologs, is that a method disclosed for producing one would provide those skilled in the art with a method for producing the other. \* \* \* Failure of the prior art to disclose or render obvious a method for making any composition of matter, whether a compound or a mixture of compounds like a zeolite, precludes a conclusion that the composition would have been obvious.")

Appellant avers that he has clearly met the burden of proof set forth in the court decisions referenced above. As stated in *In re Wiggins*, "evidence showing that attempts to prepare that compound were unsuccessful before the date of invention **will be adequate to show inoperability**" [emphasis added]. Applicant's objective evidence that 2,4-diaminopentane, the C<sub>5</sub>-diamine needed to obtain the dyes shown in prophetic Examples 48 and 74 of Harms, cannot be made by procedures generally employed in the art of amine preparation, stands unrebutted by the examiner. Indeed the Advisory Action mailed on December 8, 1999 states that Applicants arguments concerning the 2,4-diaminopentane bridge are persuasive.

However the examiner maintains that the comparison of one claimed 5 carbon bridge member compound in the Müller declaration to a 3 carbon bridge member compound of Harms does not establish the patentability of any of the three alkylene bridge member compounds in claim 16, particularly since Harms teaches advantage to 5 and 6 carbon isomers. The examiner's position appears to be that the comparison is too remote to extrapolate the results to the claimed alkylene bridge members. In this regard it has been previously noted that the inventive compound has a 3 carbon chain between the aromatic rings, with 2 methyl groups on one of the carbons. The reference compound also has a 3 carbon chain between the aromatic rings, with one methyl group on one of the nitrogens. Hence the comparison is closer than it might first appear. No 6 carbon bridges are claimed.

Additionally it is well established that comparisons need only be to compounds which are actually taught in a reference, not to compounds not exemplified. See *Ex parte Westphal et al.*, 223 USPQ 630 (PTO BOA, 1983). In this decision the Board stated at 633, "[the] latter compounds are not

exemplified ... and appellants were fully justified in testing the closest compounds actually taught in the reference".

As Judge Newman commented in *re Geiger*, 2 USPQ2d 1276, 1279 (CAFC 1987):

Applicant compared his system with the most relevant prior art. It is not required that the claimed invention be compared with subject matter that does not exist in the prior art. The applicant is not required to create prior art, nor to prove that his invention would have been obvious if the prior art were different than it actually was.

The closest taught and enabled embodiment of the reference has in fact been compared. Additional comparisons to prophetic compounds not actually disclosed should not be required. See also *In re Holladay et al.*, 199 USPQ 516 (CCPA, 1978).

In view of the narrow scope of even the broadest of the instant claims with regard to the critical component (B<sub>1</sub>), it is asserted that the objective evidence relied on is reasonably commensurate in scope with the claims. It is therefore asserted that the showing of record is sufficient to render the claims as a whole unobvious.

It is further noted that the dependent claims are necessarily narrower in scope as noted *supra*. Thus, claims 8, 9, 10 and 19 are all limited to B<sub>1</sub> as the specific alkylene of the showing. Surely these claims are reasonably commensurate in scope with the showing.

In light of the above remarks Appellant avers that the rejection of rejection of claims 2, 3, 5-10 and 16-19 under 35 U.S.C. § 103(a) as being unpatentable over Harms, GB 2,034,731 is in error as to fact and law and should also be REVERSED.

Respectfully submitted,



Kevin T. Mansfield  
Agent for Appellants  
Reg. No. 31,635

Ciba Specialty Chemicals Corp.  
Patent Department  
540 White Plains Road  
P.O. Box 2005  
Tarrytown, NY 10591-9005  
(914) 785-7127

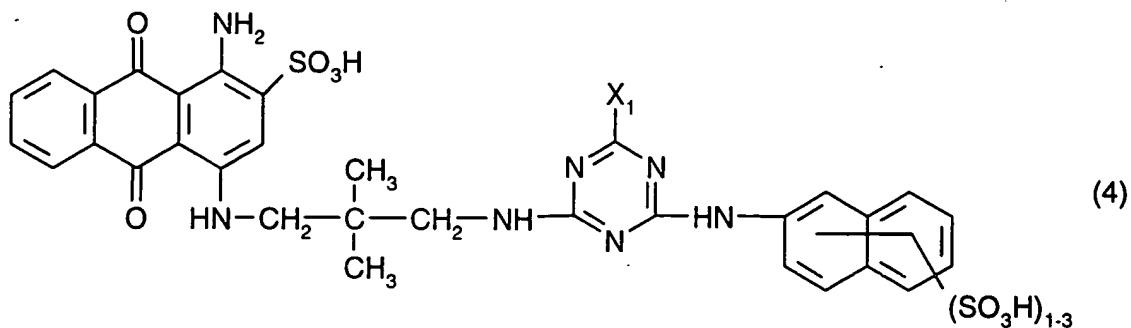
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Attachments: Fee Letter, Appendix, Claims on Appeal; 2 declarations

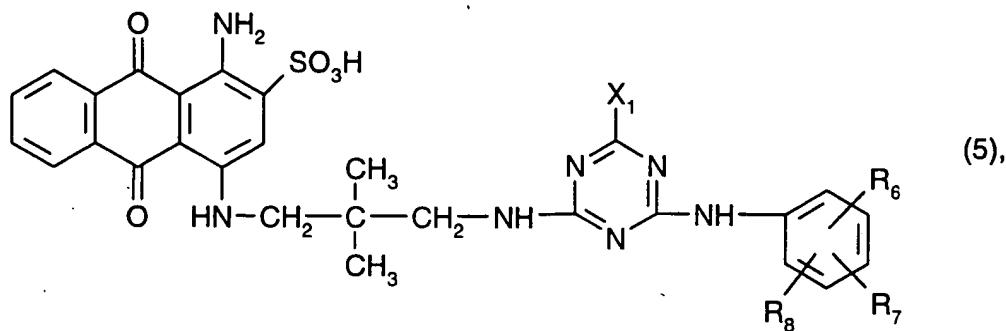
## APPENDIX

The claims on appeal are:

2. An anthraquinone dye according to claim 16, wherein R<sub>1</sub> and R<sub>2</sub> are hydrogen or C<sub>1</sub>-C<sub>4</sub>alkyl, and R<sub>3</sub> is hydrogen or C<sub>1</sub>-C<sub>8</sub>alkyl which is unsubstituted or substituted by hydroxyl, sulfo or sulfato and, with the exception of methyl, may be interrupted by oxygen.
3. An anthraquinone dye according to claim 16, wherein R<sub>1</sub>, R<sub>2</sub> and R<sub>3</sub> are hydrogen.
5. An anthraquinone dye according to claim 16, wherein Y is phenyl or naphthyl, each unsubstituted or substituted by C<sub>1</sub>-C<sub>4</sub>alkyl, C<sub>1</sub>-C<sub>4</sub>alkoxy, halogen, sulfo or a radical of formula -SO<sub>2</sub>-Z, wherein Z is a group of formula -CH=CH<sub>2</sub> or -CH<sub>2</sub>-CH<sub>2</sub>-U<sub>1</sub>, and U<sub>1</sub> is chloro or sulfato.
6. An anthraquinone dye according to claim 16, wherein B<sub>1</sub> is methylene-phenylene-methylene which is unsubstituted or substituted in the phenylene ring by C<sub>1</sub>-C<sub>4</sub>alkyl, C<sub>1</sub>-C<sub>4</sub>alkoxy, C<sub>2</sub>-C<sub>4</sub>alkanoylamino, halogen, carboxy or sulfo, or is a radical of formula -CH<sub>2</sub>-C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>-.
7. An anthraquinone dye according to claim 6, wherein B<sub>1</sub> is methylene-phenylene-methylene or a radical of formula -CH-C(CH<sub>3</sub>)-CH<sub>2</sub>-.
8. An anthraquinone dye according to claim 7, wherein B<sub>1</sub> is a radical of formula -CH<sub>2</sub>-C(CH<sub>3</sub>)<sub>2</sub>-CH<sub>2</sub>-.
9. An anthraquinone dye according to claim 16, of formula



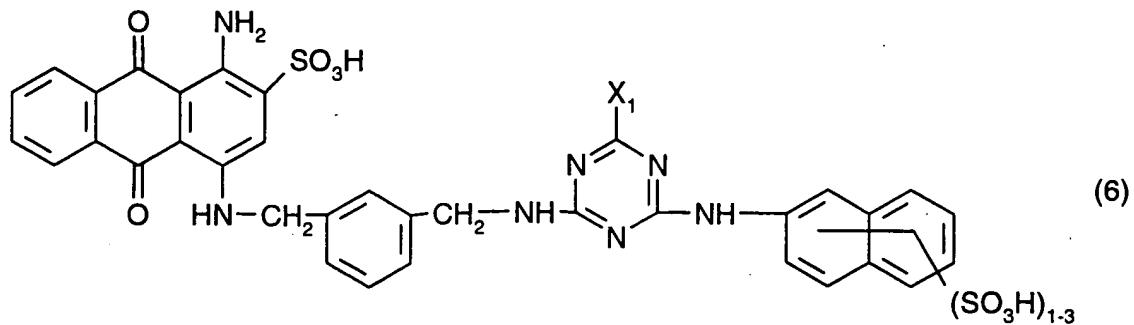
or



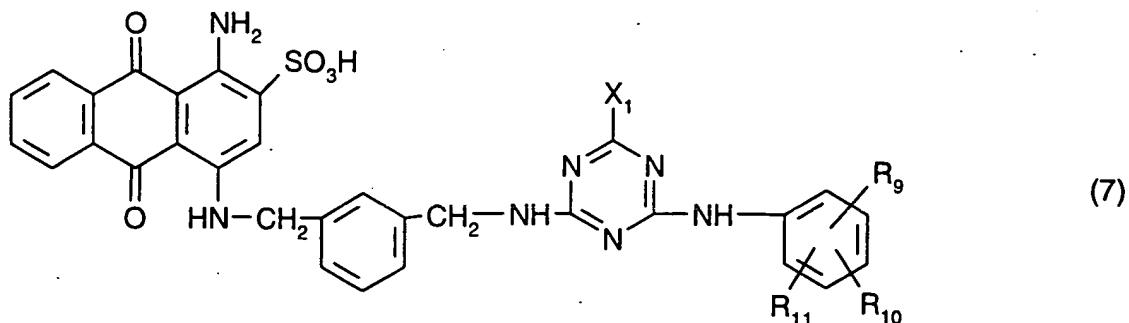
wherein  $R_6$ ,  $R_7$  and  $R_8$  are each independently of one another hydrogen,  $C_1-C_4$ alkyl,  $C_1-C_4$ alkoxy, halogen, sulfo or a radical of formula  $-SO_2-Z$ , wherein  $Z$  is a group of formula  $-CH=CH_2$  or  $-CH_2-CH_2-U_1$ , and  $U_1$  is chloro or sulfato.

10. An anthraquinone dye of formula (5) according to claim 9, wherein  $R_6$ ,  $R_7$  and  $R_8$  are each independently of one another hydrogen or sulfo.

11. An anthraquinone dye of formula



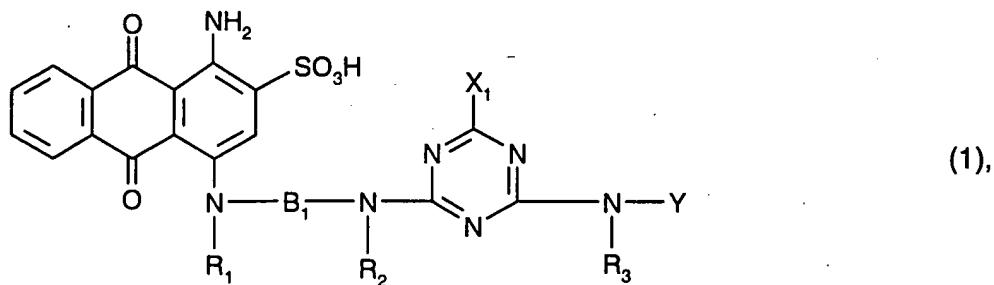
or



wherein  $R_9$ ,  $R_{10}$  and  $R_{11}$  are each independently of one another hydrogen,  $C_1$ - $C_4$ alkyl,  $C_1$ - $C_4$ alkoxy, halogen, sulfo or a radical of formula  $-SO_2-Z$ , wherein  $Z$  is a group of formula  $-CH=CH_2$  or  $-CH_2-CH_2-U_1$ , and  $U_1$  is chloro or sulfato.

12. An anthraquinone dye of formula (7) according to claim 11, wherein  $R_9$ ,  $R_{10}$  and  $R_{11}$  are each independently of one another hydrogen or sulfo.

16. An anthraquinone dye of the formula



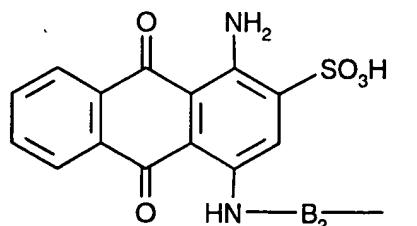
wherein

$R_1$ ,  $R_2$  and  $R_3$  are each independently of one another hydrogen or  $C_1$ - $C_{12}$ alkyl which is unsubstituted or substituted by hydroxyl, sulfo or sulfato and, with the exception of methyl, may be interrupted by oxygen,

$X_1$  is chloro or fluoro,

$B_1$  is methylene-phenylene-methylene which is unsubstituted or substituted in the phenylene ring by  $C_1-C_4$ alkyl,  $C_1-C_4$ alkoxy,  $C_2-C_4$ alkanoylamino, halogen, carboxy or sulfo, or is a radical of formula  $-CH_2-CH_2-CH(C_2H_5)-$ ,  $-CH_2-CH(OH)-CH_2-$  or  $-CH_2-C(CH_3)_2-CH_2-$ ,

$Y$  is hydrogen, or  $C_1-C_{12}$ alkyl which is unsubstituted or substituted by hydroxyl, sulfo or sulfato and, with the exception of methyl, may be interrupted by oxygen, or phenyl or naphthyl, each unsubstituted or substituted by  $C_1-C_4$ alkyl,  $C_1-C_4$ alkoxy,  $C_2-C_4$ alkanoylamino, halogen, carboxy, sulfo or a radical of formula  $-SO_2-Z$ , wherein  $Z$  is a group of formula  $-CH=CH_2$  or  $-CH_2-CH_2-U_1$ , and  $U_1$  is a leaving group, or  $Y$  is an anthraquinone of the formula

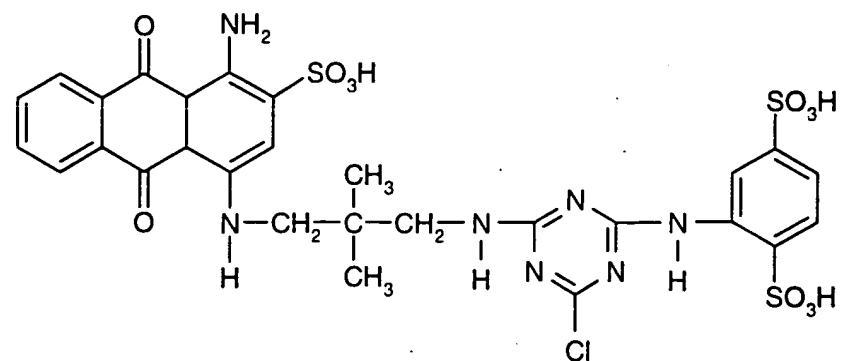


wherein  $B_2$  is  $C_2-C_{12}$ alkylene which is unsubstituted or substituted by hydroxyl, sulfo or sulfato, and which may be interrupted by oxygen, or methylene-phenylene-methylene which is unsubstituted or substituted in the phenylene ring by  $C_1-C_4$ alkyl,  $C_1-C_4$ alkoxy,  $C_2-C_4$ alkanoylamino, halogen, carboxy or sulfo.

17. A method of dyeing or printing a hydroxyl-group-containing or nitrogen-containing fibre material, which comprises the step of applying to said fibre material a tinctorially effective amount of an anthraquinone dye according to claim 16.

18. A method according to claim 17 wherein the fibre material is a cellulosic fibre material or a natural or synthetic polyamide fibre material.

19. An anthraquinone dye according to claim 9, wherein the dye of formula (5) is a dye of formula



(10).

1-20161/A

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Bernhard Müller

Serial No. 08/541,009

Group Art Unit: 1105

Filed October 11, 1995

For FIBRE-REACTIVE ANTHRAQUINONE DYES,  
PROCESS FOR THEIR PREPARATION  
AND THE USE THEREOF

Examiner: M. Einsmann

DECLARATION

I, Bernhard Müller, chemist, Doctor of Natural Science (Chemistry) from the University of Heidelberg, West Germany, a citizen of West Germany, residing at Efringen-Kirchen, West Germany, hereby declare:

That I have been employed by Ciba-Geigy as a research chemist in the Textiles Dyestuff Department since 1990;

That I have been engaged in the preparation of dyestuffs for Ciba-Geigy since 1990.

That based on the above education and experience, I consider myself an expert in the field of dyestuffs.

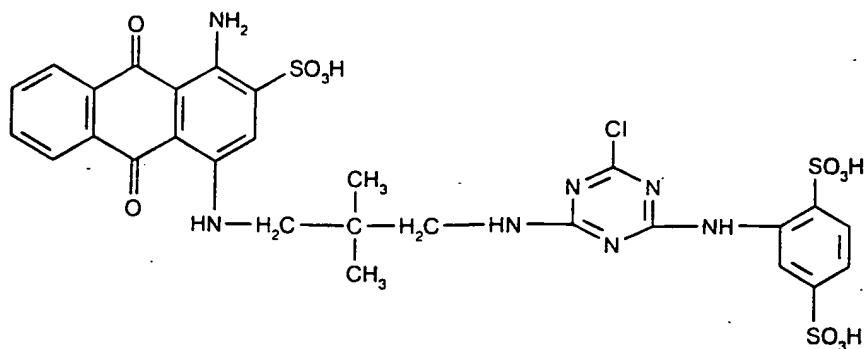
I, Bernhard Müller, declare that the preparation of dyes mentioned below and the following printings and tests were carried out under my direction and supervision.

That I am submitting herewith the following exact report of the preparation and investigation of the dyes mentioned below.

**1.) Preparation of Dyestuffs**

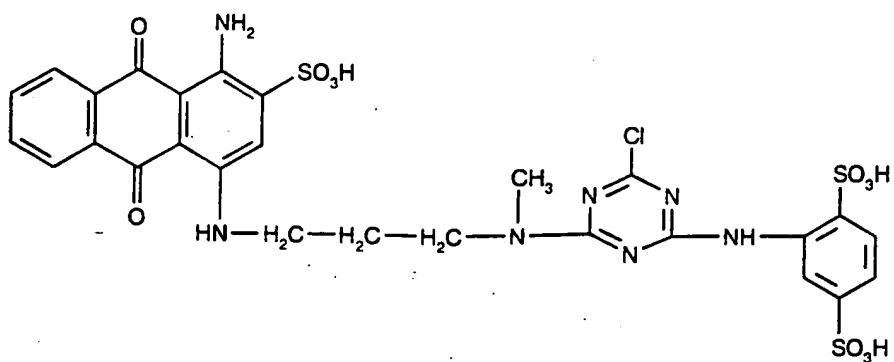
The following dyes were prepared:

Dyestuff A, the dye of formula



according to the application, supra, obtained in accordance with the instructions given in Example 78 of GB-A-2,034,731.

Dyestuff X, the dye of formula



according to GB-A-2,034,731, obtained in accordance with the instructions given in Example 78 of GB-A-2,034,731.

I, Bernhard Müller, declare that the above prepared Dyestuffs A and X exhibited the same relative purity.

**2.) Printing of mercerized cotton**

5, 15, 30, 45, 60 or 80 g of Dyestuff A (or the same amounts of Dyestuff X, respectively) were sprinkled into 500 g of a stock thickener comprising 6 % sodium alginate thickener, 20 g sodium bicarbonate, 10 g sodium m-nitro-benzenesulfonate and 3 g of a defoamer and then the printing paste in each case was adjusted by addition of water to a weight of 1000 g. Thus, for each of Dyestuffs A and X six printing pastes were prepared.

A mercerized cotton fabric was printed with the printing pastes thus obtained and dried and the resulting printed material was steamed in saturated steam at 102 °C for 4 minutes. The printed fabric was then rinsed and dried in an usual manner.

**3.) Build-up capacity**

The tinctorial strength of the printings obtained according to 2.) with 5, 15, 30, 45, 60 and 80 g of Dyestuffs A and X were measured photospectrometrically. The results are shown in Table 1 below wherein the tinctorial strength of the printings is given in units of the standard depth according to ISO 105/A-1984(E), page 4.

**Table 1 (standard depth)**

Amount of dyestuff in g	Dyestuff A standard depth	Dyestuff X standard depth
5	0.06	0.03
15	0.17	0.10
30	0.34	0.17
45	0.49	0.21
60	0.62	0.23
80	0.75	0.22

The above Table 1 shows that the inventive Dyestuff A produces strong printings in each case amounting to a maximum standard depth of about 0.75/1. In strong contradistinction thereto, the prior art Dyestuff X builds up only poorly and yields a maximum standard depth of about 0.23/1 only.

Evidently, Dyestuff A, according to the application, supra, has a much better affinity to mercerized cotton material in the printing process than prior art Dyestuff X.

I, Bernhard Müller, further declare:

That the extent of improvement attainable with Dyestuff A when compared with Dyestuff X is very surprising to me and I never would have predicted such considerable differences in the properties as shown in the Declaration especially if one concedes the structural similarity of the compounds having been tested;

That the advantages achieved with Dyestuff A are very significant ones which are of considerable practical and commercial importance. Thus it is a requirement for a dyestuff for printing cotton that it builds-up well and is suitable to print cotton in deep shades.

I, Bernhard Müller, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 Title 18 of the United States Code and that such wilful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 22<sup>nd</sup> day of August 1996

Bernhard Müller

Bernhard Müller

1-20161/A/CONT 4

July 1999

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of :  
Bernhard Müller :  
Serial No. 08/801,327 : Group Art Unit 1751  
Filed February 18, 1997 :  
For FIBRE REACTIVE ANTHRAQUINONE : Examiner M. Einsmann  
DYES, PROCESS FOR THEIR  
PREPARATION AND THE USE THEREOF

DECLARATION

I, Bernhard MÜLLER, a citizen of Germany, residing at 79588 Efringen-Kirchen, Im Mühlegrund 20, hereby declare:

That I was awarded the degree of a Doctor of Natural Science (Chemistry) of the University of Heidelberg (Germany) in 1990;

That I have been employed by Ciba Specialty Chemicals, Basel, as a chemist since 1990 and presently hold the position of a Research Chemist in the Colors Department;

That I have been engaged in the field of dyestuffs for Ciba Specialty Chemicals since 1990;

That based on the above education and experience, I consider myself an expert in the field of dyestuffs.

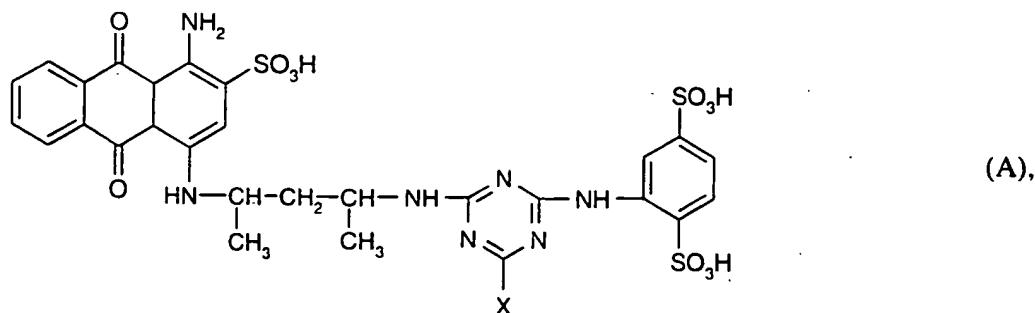
I, Bernhard MÜLLER, declare that the procedures described below were carried out under my direction and supervision:

That I am submitting herewith the following exact report of the procedures mentioned below.

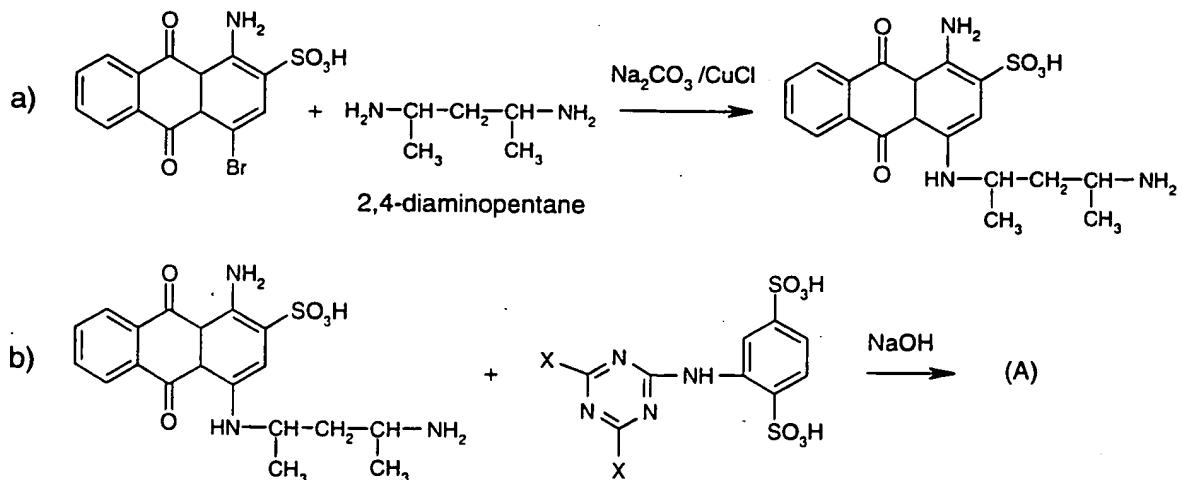
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1. Introduction

Following the instructions disclosed in Examples 36 and 73 of GB-A-2 034 731, the prior art dyes of formula (A)



wherein X is F (Example 48 of GB-A-2 034 731) or Cl (Example 74 of GB-A-2 034 731) can be obtained according to a) and b) of the following scheme, wherein X has the meanings indicated above:



GB-A-2 034 731 gives no information on the preparation of 2,4-diaminopentane used in the Ullmann-condensation outlined in a) of the scheme, supra.

2. Preparation of 2,4-diaminopentane according to procedures known in the art

2.1 Catalytic hydrogenation of pentanone-2,4-dioxime

Pantanone-2,4-dioxime was subjected to catalytic hydrogenation according to procedures A, B and C, the particulars (catalyst, pressure, work-up) and the results of which are given below:

2.1.1 Procedure A:

*catalyst:* methanol / ammonia / Raney-Nickel

*H<sub>2</sub>-pressure:* 150 bar

*work-up:* the catalyst was removed from the crude reaction mixture by filtration; volatile components were removed by distillation; the oily residue was fractionated into two fractions by distillation each of which was analysed by gas-chromatography

*result:* numerous products were detected in each fraction

2.1.2 Procedure B:

*catalyst:* ethanol / HCl / platinum oxide

*H<sub>2</sub>-pressure:* 150 bar

*work-up:* the catalyst was removed from the crude reaction mixture by filtration; volatile components were removed by distillation; the residue was made alkaline and extracted with ethyl acetate; the isolated organic phase was dried and evaporated; the residue was analysed by gas-chromatography

*result:* numerous products were detected

2.1.3 Procedure C:

*catalyst:* acetic acid / platinum oxide

*H<sub>2</sub>-pressure:* 150 bar

*work-up:* the catalyst was removed from the crude reaction mixture by filtration; volatile components were removed by distillation; the residue was made acidic by the addition of HCl and stored at 0°C (no crystallisation occurred); then the acidic residue was made alkaline by the addition of sodium hydroxide and extracted with ethyl acetate; the isolated organic phase was dried and evaporated; the residue was analysed by gas-chromatography

*result:* numerous products were detected

## 2.2 Reductive amination of 2,4-dioxopentane (acetylacetone)

Acetylacetone was added to a saturated solution of ammonia in methanol and the mixture obtained was subjected to catalytic hydrogenation under the following conditions:

catalyst: Raney-Nickel

H<sub>2</sub>-pressure: 150 bar

temperature: 50°C

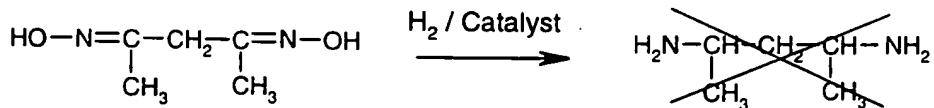
work-up: the catalyst was removed from the crude reaction mixture by filtration; volatile components were removed by distillation; the oily residue was fractionated into two fractions by distillation each of which was analysed by gas-chromatography

result: no product was detected (2,2-dimethylpropane-1,3-diamine was used as an internal reference); furthermore no blue colouring was observed once the isolated fractions were subjected to an Ullmann-condensation with 1-amino-4-bromoanthraquinone-2-sulfonic acid according to a) given in the scheme of paragraph 1. (however, the reaction mixture turned blue spontaneously when 2,2-dimethylpropane-1,3-diamine was used as a reference in an Ullmann-condensation)

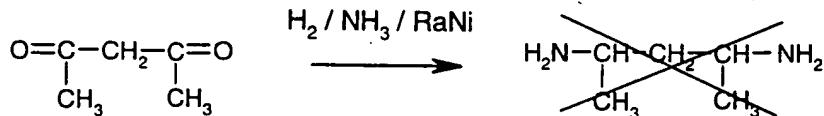
### 3. Conclusion:

- a) A uniform reaction product was not obtained by catalytic hydrogenation of pentanone-2,4-dioxime under the conditions outlined in paragraph 2.1 as revealed by gas-chromatography.

Schematically this is summarised below:



- b) Reductive amination of 2,4-dioxopentane did not yield 2,4-diaminopentane as revealed by gas-chromatography using 2,2-dimethylpropane-1,3-diamine as an internal reference. This conclusion is further supported by the fact that no blue reaction products were obtained by Ullmann-condensation. Schematically this is summarised below:



I, Bernhard MÜLLER, hereby declare:

1. That based on my education and experience, I consider myself an expert in the field of dyestuff preparation;
2. That the procedures described, supra, and the conditions thereof are known in the art of amine preparation and are typically applied for that purpose;
3. That the results show that 2,4-diaminopentane was not obtained by the procedures described, supra.

I, Bernhard MÜLLER, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that wilful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 Title 18 of the United States Code and that such wilful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 7th day of July 1999



Bernhard MÜLLER